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## FACSIMILE COVER SHEET

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DATE: September 20, 2006

TO: Examiner Edward M. Johnson  
Group Art Unit 1754

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Application No.: 09/757,519

OUR REF.: 3132.07US02

Applicant: Horne et al.

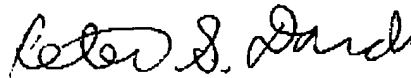
Due Date: September 22, 2006

FROM: Peter S. Dardi, Ph.D.  
PHONE #: 404-949-5730

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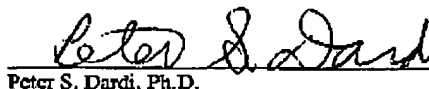
- (1) Appeal Brief Transmittal; and
- (2) Appeal Brief with appendices.

Respectfully submitted,

Peter S. Dardi, Ph.D.  
Registration No. 39,650

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Attorney Docket No. 3132.07US02

## APPEAL BRIEF TRANSMITTAL

In re the application of:

	Horne et al.	Confirmation No.: 3132.07US02
Application No.:	09/757,519	Examiner: Johnson, Edward M.
Filed:	January 9, 2001	Group Art Unit: 1754
For:	METAL VANADIUM OXIDE PARTICLES	

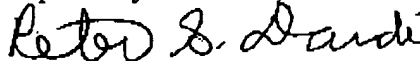
Mail Stop Appeal Brief-Patents  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Transmitted herewith, is the Appeal Brief in the above-identified application, with respect to the Notification of Non-Complaint Appeal Brief filed on August 20, 2003.

Applicants previously paid \$250 for the filing of an Appeal Brief in this matter on August 20, 2003. Applicants believe that no additional fees are due, but please charge the below deposit account if that assessment is in error to ensure entry of this Brief.

Respectfully submitted,



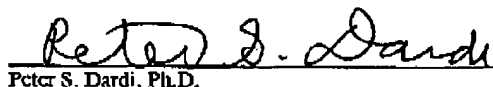
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Peter S. Dardi, Ph.D.

SEP 20 2006

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

Attorney Docket No.: 3132.07US02

Horne et al.

Confirmation No.: 8679

Application No.: 09/757,519

Examiner: Edward M. Johnson

Filed: January 9, 2001

Group Art Unit: 1754

For: METAL VANADIUM OXIDE PARTICLES

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BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

BRIEF FOR APPELLANT

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SEP 20 2006

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Application No. 09/969,025

PATENT APPLICATION  
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

Attorney Docket No.: 3132.07US02

Horne et al.

Confirmation No.: 8679

Application No.: 09/757,519

Examiner: Edward M. Johnson

Filed: January 9, 2001

Group Art Unit: 1754

For: METAL VANADIUM OXIDE PARTICLES

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BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES  
APPEAL BRIEFMail Stop Appeal Brief - Patents  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

INTRODUCTORY COMMENTS

This is an appeal from an Office Action dated January 27, 2003, in which claims 1-3, 6-10, 17 and 22-26 were finally rejected. A Notice of Appeal was filed on June 23, 2003. An Appeal Brief was filed on August 20, 2003. A Notice of Non-Compliant Amendment was mailed on August 22, 2006. This Corrected Appeal Brief is timely filed in response to the Notice of Non-Compliant Brief.

*Please grant any extension of time necessary for entry; charge any fee due to Deposit Account No. 50-3863.*

## CERTIFICATE OF MAILING

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Peter S. Dardi, Ph.D.

SEP 20 2006

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Application No. 09/969,025

**REAL PARTY IN INTEREST**

Greatbatch, Inc. (previously known as Wilson Greatbatch Technologies, Inc.), has acquired the entire right, title and interest in and to the invention, the application, and any and all patents to be obtained therefore. Greatbatch was assigned the patent application by NanoGram Corporation, a corporation organized under the laws of the state of Delaware, and having offices at 165 Topaz St., Milpitas, California. On June 17, 2004, an assignment was executed transferring ownership from NanoGram Corporation to Wilson Greatbatch Technologies, Inc., although NanoGram Corp. may have retained certain licensed rights, recorded at reel 015552, frame 0199. The rights in the patent application were transferred to NanoGram Corp. as per the Assignment, recorded at Reel 011451, Frame 0570 from the inventors to NeoPhotonics Corporation and an assignment from NeoPhotonics Corporation to NanoGram Corporation recorded at Reel 013957, Frame 0076. Note that NeoPhotonics Corporation was formerly called NanoGram Corporation, and the present NanoGram Corporation is an independent corporation spun out from NeoPhotonics Corp.

**RELATED APPEALS AND INTERFERENCES**

Pending U.S. Patent application 09/606,884 was appealed to the USPTO Board of Patent Appeals and Interferences. This patent application is assigned to NanoGram Corporation. A copy of the decision is attached. It is noted that the decision reversed the rejection.

**STATUS OF CLAIMS**

Claims 1-3, 6-18 and 22-26 are pending, and claims 4, 5 and 19-21 have been canceled. Claims 1-3, 6-10, 17 and 22-26 stand rejected. Claims 11-16 and 18 are free of any rejections and are objected to for depending on a rejected base claim. The pending claims are listed in the Claims Appendix.

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Application No. 09/969,025

**STATUS OF AMENDMENTS**

All Amendments have been entered with the filing of the Appeal.

**SUMMARY OF CLAIMED SUBJECT MATTER**

The invention relates to powders, i.e., collections of particles, having a composition of a metal vanadium oxide. Metal vanadium oxides have a non-vanadium metal ion along with a vanadium ion within an oxide composition. The claimed composite metal oxide particles have an average particle size less than a micron.

Any particular powder has particles that can be characterized by size. A collection of particles has an average particle size and a distribution of particle sizes, which are related but separate properties. The distribution of particle sizes relate to the size uniformity of the particles. Some of the pending claims specify particular distributions corresponding to highly uniform particles. All of the claims directed to particle collections have a submicron average particle size.

Some of the pending claims relate to methods for forming metal vanadium oxide particles. In the claimed methods, the metal vanadium oxide particles are formed by heating a mixture of vanadium oxide particles with a non-vanadium metal compound. The reactant vanadium oxide particles have an average particle size less than a micron. Applicants' specification describes the formation of submicron vanadium oxide particles using a process called laser pyrolysis. Through the description of the laser pyrolysis approach, Applicants' specification enables the formation of the starting materials for the formation of submicron metal vanadium oxide materials. The present application does not claim the formation of particles with laser pyrolysis.

Additional claims are directed to batteries formed with submicron metal vanadium oxide particles. In particular, metal vanadium oxide particles are useful as cathode materials, especially

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for lithium-based batteries. The submicron character of the metal vanadium oxide particles can contribute improved performance in battery applications.

**GROUND OF REJECTION TO BE REVIEWED ON APPEAL**

1. The rejection of claims 1-3, 6-9, 17 and 22-26 as unpatentable under 35 U.S.C. § 102(b) as anticipated by U.S. Patent 5,549,880 to Koksang.
2. The rejection of claim 10 as unpatentable under 35 U.S.C. § 102(b) as anticipated by U.S. Patent 5,512,214 to Koksang?

**ARGUMENT**

**GROUPING OF CLAIMS**

The following argument is organized around the following grouping of claims, which is summarized for the convenience of the Board.

1. Claims 1, 17, 22, 24 and 26 are within a first claim group directed to a collection of metal vanadium oxide particles or batteries formed with metal vanadium oxide particles with the particles having a specified average particles size.
2. Claim 2 is in a second claim group directed to a collection of metal vanadium oxide particles with an average diameter from about 5 nm to about 100 nm.
3. Claim 3 is in a third claim group directed to a collection of metal vanadium oxide particles with an average diameter from about 5 nm to about 50 nm.

4. Claims 6-9 and 23 are within a forth claim group directed to a collection of metal vanadium oxide particles or batteries formed with metal vanadium oxide particles with the particles having a specified average particles size and a specified particle size uniformity.

5. Claims 10 and 25 are within a fifth claim group directed to a method for producing metal vanadium oxide particles using vanadium oxide particles with a specified average particle size range.

#### LEGAL AUTHORITY

The Court of Appeals for the Federal Circuit has exclusive appellate jurisdiction for cases arising under the patent law under 28 U.S.C. § 1295 (a)(1). The Federal Circuit has adopted as binding precedent all holding of its predecessor courts, the U.S. Court of Claims and the U.S. Court of Customs and Patent Appeals. South Corp. v. U.S., 215 USPQ 657 (Fed. Cir. 1982). Therefore, unless they have been overruled en banc, CCPA cases are binding precedent for the present appeal.

#### A. ANTICIPATION

##### 1. A Single Reference Must Disclose Every Element Set Forth In a Claim To Anticipate The Claim

"For a prior art reference to anticipate in terms of 35 U.S.C. § 102, every element of the claimed invention must be identically shown in a single reference. **These elements must be arranged as in the claim under review**, but this is not an 'ipsissimis verbis' test." In re Bond, 15 USPQ2d 1566, 1567 (Fed. Cir, 1990)(Internal citations omitted and emphasis added.).



"If the prior art reference does not expressly set forth a particular element of the claim, that reference still may anticipate if that element is 'inherent' in its disclosure. To establish inherency, the intrinsic evidence 'must make it clear that the missing descriptive matter is necessarily present in the thing described in the reference, and that it would be so recognized by persons of ordinary skill. Inherency, however, may not be established by probabilities or possibilities. The mere fact that a certain thing may result from a given set of circumstances is not sufficient.'" In re Robertson, 49 USPQ2d 1949, 1950, 1951 (Fed. Cir. 1999), citing Continental Can Co. v. Monsanto Co., 20 USPQ2d 1746, 1749 (Fed. Cir. 1991).

"Every element of the claimed invention must be literally present, arranged as in the claim. The identical invention must be shown in as complete detail as is contained in the patent claim." Richardson v. U.S. Suzuki Motor Corp., 9 USPQ2d 1913, 1920 (Fed. Cir. 1989)(Internal citations omitted, and emphasis added.); see also MPEP 2131. "Here, as well, anticipation is not shown by a prior art disclosure which is only 'substantially the same' as the claimed invention." Jamesbury Corp. v. Litton Industrial Products, Inc., 225 USPQ 253, 256 (Fed. Cir. 1985)(emphasis added).

## 2. Ranges

Claims covering a range of composition narrower than a broader range covered in the prior art are not anticipated, although they may be obvious over the prior art. In re Malagari, 182 USPQ 549, 553 (CCPA 1974). Such claims are analogous to the claim of a species or subgenus within a genus, which may be patentable and generally are not obvious. See MPEP 2131.02 and 2131.03.

## B. OBVIOUSNESS

### 1. The Examiner Bears The Burden Of Demonstrating Obviousness.

The Applicants note that the patent office has the burden of persuasion in showing that the Applicants are not entitled to a patent. "[T]he conclusion of obviousness vel non is based on the

preponderance of evidence and argument in the record." In re Oetiker, 24 USPQ2d 1443, 1445 (Fed. Cir. 1992). The patent office has the ultimate burden of persuasion in establishing that an applicant is not entitled to a patent. Id. at 1447, concurring opinion of Judge Plager. "The only determinative issue is whether the record as a whole supports the legal conclusion that the invention would have been obvious." Id.

"In rejecting claims under 35 U.S.C. §103, the examiner bears the initial burden of presenting a prima facie case of obviousness." In re Rijckaert, 28 USPQ2d 1955, 1956 (Fed. Cir. 1993). If the Examiner fails to establish a prima facie case of obviousness, the obviousness rejection must be withdrawn as a matter of law. In re Ochiai, 37 USPQ at 1131 ("When the references cited by the examiner fail to establish a prima facie case of obviousness, the rejection is improper and will be overturned"). "If examination at the initial stage does not produce prima facie case of unpatentability, then without more the applicant is entitled to grant of the patent." In re Oetiker, 24 USPQ2d 1443, 1444 (Fed. Cir. 1992).

"Only if that burden is met, does the burden of coming forward with evidence or argument shift to the applicant." In re Rijckaert, 28 USPQ2d at 1956. "After evidence or argument is submitted by the applicant in response to an obviousness rejection, 'patentability is determined on the totality of the record, by a preponderance of the evidence with due consideration to persuasiveness of the argument.'" In re Chu, 36 USPQ2d 1089, 1094 (Fed. Cir. 1995)(quoting In re Oetiker, 24 USPQ2d 1443, 1444 (Fed. Cir. 1992)).

## 2. The References Must Teach Or Suggest All Of The Claim Elements

Prima facie obviousness is not established if all the elements of the rejected claim are not disclosed or suggested in the cited art. In re Ochiai, 37 USPQ 1127, 1131 (Fed. Cir. 1995). ("The test for obviousness *vel non* is statutory. It requires that one compare the claim's 'subject matter as a whole' with the prior art 'to which said subject matter pertains.'"). See also, MPEP 2143.03 "All

Claim Limitations Must Be Taught or Suggested," citing In re Royka, 180 USPQ 580 (CCPA 1974). "To establish prima facie obviousness of a claimed invention, all of the claim limitations must be taught or suggested by the prior art." MPEP 2143.03.

To establish prima facie obviousness, all the elements of the claim must be taught or suggested by the cited references without the benefit of hindsight based on the applicant's own disclosure. "To imbue one of ordinary skill in the art with knowledge of the invention in suit, when no prior art reference or references of record convey or suggest that knowledge, is to fall victim to the insidious effect of a hindsight syndrome wherein that which only the inventor taught is used against its teacher." W. L. Gore & Assocs., Inc. v. Garlock, Inc., 220 USPQ 303, 312-13 (Fed. Cir. 1983). "Skill in the art does not act as a bridge over gaps in the substantive presentation of an obviousness case, but instead supplies the primary guarantee of objectivity in the process." All-Site Corp. v. VSI International Inc., 50 USPQ2d 1161, 1171 (Fed. Cir. 1999).

### 3. The Claimed Recited Properties Of The Claimed Compositions Of Matter Are Claim Elements

It is long established that a composition of matter is indistinguishable from its properties. In re Papesch, 137 USPQ 43, 51 (CCPA 1963); In re Cescon, 177 USPQ 264, 266 (CCPA 1973). There are two types of properties, chemical/compositional properties and physical properties. The chemical/compositional properties of the composition of matter determine what the material is, while the physical properties relate to the interaction and behavior of the composition of matter. Often unique or unexpected physical properties are used to establish the existence of an unobvious composition when chemical/compositional properties either are unknown or do not fully represent the unobviousness of the composition. However, discovery of a surprising or unexpected physical property does not necessarily control an obviousness determination, and all the evidence under the Graham factors must be considered. See, for example, Richardson-Vicks v. Upjohn Co., 44

USPQ2d 1181, 1187 (Fed. Cir. 1997). **In the present case, the claims do not relate to the discovery of properties of previously known or suggested materials.**

Obviousness under 35 U.S.C. §103 must be evaluated by viewing the invention as a whole. In re Langer, 175 USPQ 169, 171 (CCPA 1972). "In effect, we consider the prior art 'as a whole' with the claimed subject matter 'as a whole.'" Id. This rule superseded other principles, and specifically, "homology should not be automatically equated with prima facie obviousness." Id. (emphasis added). "To give meaning to the language of 35 U.S.C. 103 which speaks to the subject matter 'as a whole,' we feel weight must be given the properties of a compound or composition of matter." In re Murch, 175 USPQ 89, 92 (CCPA 1972)(emphasis added).

The present claims are directed to compositions of matter or method for manipulating compositions of matter. Certain claimed aspects of the present invention are chemical/compositional properties that make the material a different composition of matter. In particular, Applicants' **claimed compositions** have several compositional features of particular relevance. First, the composition of matter comprises particles with specified properties. Specifically, the particles have a specified range of average particle sizes. Average particle size is a chemical/compositional property similar to chemical formula or molecular weight of a polymer. Collections of particles with one average particle size are a different composition of matter and will have different physical properties from collections of particles with other average particle sizes.

Similarly, the distribution of particle sizes is another independent chemical/composition property of solid particles that is distinct from the average particle size. **A particle collection with a particular particle size distribution is a different composition of matter and will have different physical properties from other collections of particles with different particle size distributions.** Applicants have developed an approach using light/radiation-based pyrolysis to produce the highly uniform powders/particles, which is the subject of some of the present claims.

These highly uniform particles can be further reacted to form other product particles with desirable properties. A particle collection with a narrow particle size distribution is more uniform.

4. To Support A Finding Of Obviousness Based On Cited Art, The Cited Art Must Provide A Means Of Obtaining The Claimed Composition Or Apparatus

The proposition is well established that the cited art only renders a composition of matter or apparatus unpatentable to the extent that the cited art enables the disputed claims, in other words, if the cited art provides a means of obtaining the claimed composition or apparatus.

To the extent that anyone may draw an inference from the Von Bramer case that the mere printed conception or the mere printed contemplation which constitutes the designation of a 'compound' is sufficient to show that such a compound is old, regardless of whether the compound is involved in a 35 U.S.C. 102 or 35 U.S.C. 103 rejection, we totally disagree. ... We think, rather, that the true test of any prior art relied upon to show or suggest that a chemical compound is old, is whether the prior art is such as to place the disclosed 'compound' in the possession of the public. In re Brown, 141 USPQ 245, 248-49 (CCPA 1964)(emphasis in original)(citations omitted).

Similarly, see In re Hoeksema, 158 USPQ 596, 600 (CCPA 1968)(emphasis in original):

We are certain, however, that the invention as a whole is the claimed compound and a way to produce it, wherefore appellant's argument has substance. There has been no showing by the Patent Office in this record that the claimed compound can exist because there is no showing of a known or obvious way to manufacture it; hence, it seems to us that the 'invention as a whole,' which section 103 demands that we consider, is not obvious from the prior art of record.

While there are valid reasons based on public policy as to why this defect in the prior art precludes a finding of obviousness under section 103, In re Brown, supra, its immediate significance in the present inquiry is that it poses yet another difference between the claimed invention and the prior art which must be considered in the context of section 103. So considered, we think the differences between appellant's invention as a whole and the prior art are such that the claimed invention would not be obvious within the contemplation of 35 U.S.C. 103.

The Federal Circuit has further emphasized these issues. "But to be prior art under section 102(b), a reference must be enabling. That is, it must put the claimed invention in the hands of one skilled in the art." In re Sun, 31 USPQ2d 1451, 1453 (Fed. Cir. 1993)(unpublished). Assertions in a prior art reference do not support an anticipation or obviousness rejection unless the references place the claimed invention in the hands of the public. Beckman Instruments Inc. v. LKB Produkter AB, 13 USPQ2d 1301, 1304 (Fed. Cir. 1989). "In order to render a claimed apparatus or method obvious, the prior art must enable one skilled in the art to make and use the apparatus or method." Id. While a properly citable reference is prior art for all that it teaches, references along with the knowledge of a person of ordinary skill in the art must be enabling to place the invention in the hands of the public. In re Paulsen, 31 USPQ2d 1671, 1675 (Fed. Cir. 1994). See also In re Donohue, 226 USPQ 619, 621 (Fed. Cir. 1985).

#### 5. Obviousness Over A Single Prior Art Reference

The importance of the principle that the prior art itself must suggest the motivation to modify the teachings of a reference was eloquently stated in In re Rouffet, 47 USPQ2d 1453, 1458 (Fed. Cir. 1998)(emphasis added):

The Board did not, however, explain what specific understanding or technical principle within the knowledge of one of ordinary skill in the art would have suggested the combination. Instead the board merely invoked the high level of skill in the field of the art. If such a rote invocation could suffice to supply a motivation to combine, the more sophisticated scientific fields would rarely, if ever, experience a patentable technical advance. Instead, in complex scientific fields, the Board could routinely identify the prior art elements in an application, invoke the lofty level of skill, and rest its case for rejection. To counter this potential weakness in the obviousness construct, the suggestion to combine requirement stands as a critical safeguard against hindsight analysis and rote application of the legal test for obviousness.

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Similar principles must be applied when obviousness is based on the teachings of a single cited reference.

In appropriate circumstances, a single prior art reference can render a claim obvious. However, there must be a showing of a suggestion or motivation to modify the teachings of that reference to the claimed invention in order to support the obviousness conclusion. This suggestion or motivation may be derived from the prior art reference itself, from the knowledge of one of ordinary skill in the art, or from the nature of the problem to be solved. **Determining whether there is a suggestion or motivation to modify a prior art reference is one aspect of determining the scope and content of the prior art, a fact question subsidiary to the ultimate conclusion of obviousness.**

Sibia Neurosciences, Inc. v. Cadus Pharmaceutical Corp., 55 USPQ2d 1927, 1931 (Fed. Circuit 2000)(internal citations omitted, emphasis added).

#### ANALYSIS

##### A. REJECTION OVER KOKSBANG '880

The Examiner rejected claims 1-3, 6-9, 17 and 22-26 under 35 U.S.C. § 102(b), as anticipated by U.S. Patent 5,549,880 to Koksang (the Koksang '880 patent). The Examiner has failed to establish a prima facie case of anticipation. Furthermore, in phone interviews, the Examiner has raised issues relating to obviousness that have not been raised in written Office Actions. The implication from these discussions was that the Patent Office is operating on the incorrect basis that Applicants have the burden to establish patentability. The Examiner has not asserted a prima facie case of obviousness. These issues are analyzed in detail in the following.

Also, Applicants note that it seems to have been an error to include claim 25 in this rejection since claim 25 depends from claim 10, which was not rejected over the Koksang '880 patent. Applicants respectfully request clarification of the status of claim 25. To the extent that claim 25 is rejected over Koksang '214 patent, the patentability of claim 25 is argued below with respect to claim 10 since the arguments apply equally with claim 25.

Group 1 Claims

The Patent Office has the burden to establish prima facie unpatentability. The rejection formally presented by the Examiner is based on anticipation. Since the Examiner has raised issued in phone interviews based on obviousness, both anticipation and obviousness are discussed separately below.

The Koksbang '880 patent does not prima facie anticipate Applicants' claimed invention. Specifically, pending claim 1 specifies that the average particle size for the claimed collection of metal vanadium oxide particles is less than a micron. The Koksbang '880 patent does not disclose metal vanadium oxide particles with an average particle size less than a micron either **explicitly or inherently**. The Koksbang '880 patent describes a lithium vanadium oxide, which is a species of metal vanadium oxides, "in the form of a fine powder having a surprisingly small particle size on the order of 0.1 to 5 microns, and typically less than 10 microns." Column 2, lines 59-61. The Koksbang patent **does not identically disclose** the composition of Applicants' invention since the Koksbang patent does not expressly recite that the particle size range disclosed is a range of average particle sizes.

Furthermore, it is clear in context that this description of particle sizes is not a description of average particle sizes. In particular, the inclusion of the description of "typically less than 10 microns" is inconsistent with 0.1 to 5 microns being a range of averages. If 0.1 to 5 microns relates to a particular distribution of particle sizes, there is some cut off in the distribution that is used to assign the end points presented. This can be picked, for example, at one standard deviation. Thus, there would be some particles with sizes larger than 5 microns and smaller than 0.1 microns. To then indicate that the particle sizes are typically less than 10 microns would imply that a more stringent criterion is being used to indicate "typically," for example, 90 percent or 95 percent of the particles. This interpretation is perfectly consistent although not explained in the Koksbang patent. So if the "0.1 to 5 microns" is a distribution, the expression "typically less



than 10 microns" is not inconsistent. However, if "0.1 to 5 microns" refers to average particle sizes, the expression "typically less than 10 microns" is inconsistent since a description clearly relating to the distribution would then be contrasted with averages without explanation. Since there is no explicit explanation otherwise, the consistent reading of the language in the Koksbang '880 patent is that all values relate to the distribution of particle sizes.

In addition, the Koksbang '880 patent has a single example directed to the production of lithium vanadium oxide. A single set of reaction conditions are described for the production of the lithium vanadium oxide from column 4, line 49 to 67. "The product was found to have a surprisingly small particle size on the order of 0.1 to 5 microns, and typically less than 10 microns." Column 5, lines 4-6. A powder product has a single average particle size and a single particle size distribution relating to the characteristics of the particles within the powder. Since the quoted language was used to describe a single product, it must be referring to a single distribution of particle sizes that would have a corresponding a single average particle size, not a range of average particle sizes. Based on a single example with one set of reaction conditions, the only consistent interpretation of the language in the Koksbang '880 patent is that 0.1 to 5 microns refers to a single distribution with an average particle size of roughly 2.5 microns. Since the single set of particle properties disclosed in the Koksbang '880 patent have an average particle size significantly greater than the claimed particle size, the Koksbang '880 patent does not explicitly disclose a collection of metal vanadium oxide particles with an average particle size less than one micron.

Similarly, the Koksbang '880 patent does not inherently disclose metal vanadium oxide particles with an average particle size less than one micron. Specifically, the Koksbang patent explicitly discloses the particle size of the metal vanadium oxide particles. There are no particle collections in the Koksbang '880 patent with inherent size properties that are not described, so there cannot be inherent disclosure relating to average particle sizes. Since the Koksbang '880

patent does not explicitly or inherently disclose metal vanadium oxide particles with an average particle size less than a micron, the Koksbang '880 patent does not anticipate Applicants' claimed invention.

With respect to obviousness, the Examiner has clearly not stated a prima facie case for obviousness. It is simply not the Applicants' burden to establish patentability. With respect to the Examiner's assertions regarding the possible formation of Applicants' claimed particle from the particles formed by the process of the Koksbang '880 patent, this can only be relevant to an obviousness analysis. However, the Koksbang '880 patent does not teach or suggest any separation techniques. Under well established legal principles, the modification of the teachings of a reference can only be based upon the teachings of another reference or what is well known to a person of ordinary skill in the art. The Examiner has not asserted that appropriate teaching is well known in the art or provided a reference that describes appropriate knowledge being well known in the art. If it was well known to a person of ordinary skill in the art how to form the claimed particle collections from the particle collections described in the Koksbang '880 patent, it should be possible for the Examiner to provide such a reference. Similarly, the Examiner has not asserted or implied in the phone conferences that he has personal knowledge that appropriate approaches are known in the art for performing the requires submicron particle separation.

Cited references must teach all of the claim elements. Specifically, the cited art must place the invention in the hands of the public to support an obviousness or anticipation rejection. The Koksbang '880 patent simply does not put the invention in the hands of the public. Certainly, with respect to Applicants' claimed invention, the Examiner has fallen far short of meeting his burden of establishing prima facie anticipation or obviousness.

The Examiner's implied assertion that Applicants must provide evidence of more with respect to showing that there is no other way of making Applicants' claimed invention is a shifting of burdens contrary to the law. Applicants simply do not as a matter of law have the

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burden to prove patentability. If a person of ordinary skill in the art could practice the claimed invention without undue experimentation based on the disclosure in the Koksbang '880 patent, the Examiner should easily be able to support that assertion with some kind of evidence. The Examiner has presented no evidence to support an obviousness rejection over the Koksbang '880 patent.

Since the Examiner has fallen short of establishing prima facie unpatentability of Applicants' claimed invention, the rejection should be withdrawn.

#### Group 2 Claim

The claims of group 2 relate to collections of metal vanadium oxide particles with an average particle size from 5 nm (0.005 microns) to 100 nm (0.1 microns). The Examiner has not clearly indicated in any Office Action how this claim is anticipated by the teachings of the Koksbang '880 patent. Therefore, the Examiner has failed to establish prima facie anticipation of this claim.

#### Group 3 Claim

The claim of group 3 relate to collections of metal vanadium oxide particles with an average particle size from 5 nm (0.005 microns) to 50 nm (0.05 microns). The Examiner has not indicated in any Office Action how this claim can possibly be anticipated by the Koksbang '880 patent since there is no description whatsoever of particles this small in the Koksbang '880 patent. Thus, the Examiner has clearly failed to assert prima facie anticipation of the Group 3 claim.

Group 4 Claims

Group 4 claims include additional features relating to the uniformity of the particles with respect to size. Specifically, the particle size distributions are narrow as specified in the claims. With respect to these claims, the Examiner asserted that these uniformity features were inherent in the Koksbang '880 particles since the Koksbang '880 patent teaches the same process. See the Office Action of January 27, 2003 at page 3. This simply is not the case. The Koksbang patent throughout teaches a solution based reaction. In contrast, Applicants method involved forming a vanadium oxide reactant that is formed in a flow with the reaction driven by an intense light source. The vanadium oxide reactant is converted into the metal vanadium oxide in a solid state reaction. Since the cited reference does not teach or suggest the same methodology as Applicants' disclosure, the Examiner has clearly failed to assert a prima facie case of anticipation of the claims of group 4.

When Applicants previously pointed out that the methods were not the same, the Examiner asserted that the claims did not specify a method. But with all due respect, the Examiner's comment is irrelevant. Applicants described their method only to refute the Examiner's assertions that the method was the same as the Koksbang '880 method and not to assert that Applicants' were relying on their process for patentability. The legal standards for establishing anticipation by inherency of a claimed composition of matter stipulate that the compositions in the cited references must necessarily have the claimed characteristics. However, there is absolutely no basis to believe that the particle collections in the Koksbang '880 patent have a uniformity disclosed and claimed by Applicants. The Koksbang '880 patent simply does not prima facie anticipate explicitly or inherently Applicants' invention in Group 4 claims.

Summary

The Examiner has failed to meet his burden of establishing prima facie unpatentability of Applicants' claimed invention based on the Koksbang '880 patent. Applicants respectfully request withdrawal of the rejection of claims 1-3, 6-9, 17 and 22-26 under 35 U.S.C. § 102(b), as anticipated by the Koksbang '880 patent.

**B. REJECTIONS OVER KOKSBANG '214**

The Examiner rejected claim 10 under 35 U.S.C. § 102(b) as being anticipated by U.S. Patent 5,512,214 to Koksbang (the Koksbang '214 patent). Claim 10 is directed to a method for producing metal vanadium oxide particles using submicron vanadium oxide particles. The Examiner asserted that the Koksbang '214 patent discloses vanadium oxide particles with an average particle size less than a micron at column 5, lines 1-6. The Koksbang '214 patent, however, does not disclose submicron vanadium oxide particles. **Based on well established case law**, the Koksbang '214 patent does not prima facie anticipate Applicants' claimed invention. Applicants respectfully request reconsideration of the rejection based on the following analysis.

The Koksbang '214 patent is directed to the synthesis of vanadium pentoxide ( $V_2O_5$ ) and lithium vanadium oxide from the vanadium pentoxide. At column 5, lines 1-4 (emphasis added), the Koksbang '214 patent indicates that "vanadium oxide particles are formed having an average size less than 100 microns, desirably less than 50 microns and, preferably, less than 10 microns." The Koksbang '214 patent does not disclose an average particle size of less than about 1 micron, as disclosed and claimed by Applicants.

It is well established that a broader range is clearly not anticipated by a narrower range. Therefore, the Koksbang '214 patent simply does not anticipate Applicants' claimed invention. The Examiner has failed to establish a case of prima facie anticipation. Also, the Examiner has failed to assert any case for obviousness. Nevertheless, issues raised by the Examiner in phone interviews

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seem to relate to obviousness. Therefore, Applicants address the obviousness issue, although the Examiner has not formulated an obviousness argument.

In order to render a claimed process obvious, the cited references must place the claimed process in the hands of the public. The Koksbang '214 patent does not disclose how to form vanadium oxide particles with an average particle size less than a micron. The range disclosed in the Koksbang '214 patent is a factor of ten greater than Applicants' claimed particle sizes. The Examiner has not indicated how such a reduction in particle size can be performed. While the Koksbang '214 patent disclosed the desirability of having smaller average particle sizes, the Koksbang '214 patent does not indicate that particles can be formed with an average particle size less than a micron is achievable. This gap with respect to smaller vanadium oxide particles in the disclosure of the Koksbang '214 patent strongly suggests that Koksbang '214 patent does not enable a person of ordinary skill in the art to form submicron vanadium oxide particles.

In summary, the Koksbang '214 patent simply does not prima facie anticipate Applicants' claimed invention. The Examiner has not indicated any basis for establishing prima facie obviousness. Applicants do not have the burden to establish patentability, although Applicants' do not see any issues within the references of record that call patentability into question.

Since a prima case for anticipation or obviousness has not been established by the Examiner, Applicants respectfully request withdrawal of the rejection of claim 10 under 35 U.S.C. § 102(b) as being anticipated by the Koksbang '214 patent.

#### Summary

Appellants' have presented clear evidence that the prior art does not place Applicants' claimed invention in the hands of the public. Since WO 00/38282 does not render Applicants' claimed invention *prima facie* obvious, Applicants respectfully request withdrawal of the

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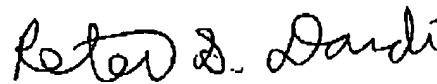
Application No. 09/969,025

rejection of claims 3, 6-10, 15 and 36-54 under 35 U.S.C. § 103(a) as obvious over WO 00/38282.

CONCLUSIONS AND REQUEST FOR RELIEF

Applicants submit that claims 1-3, 6-18 and 22-26 are in condition for allowance. Thus, Applicants respectfully request the reversal of the rejections of claims 1-3, 6-10, 17 and 22-26 and the allowance of claims 1-3, 6-18 and 22-26.

Respectfully submitted,



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## CLAIMS APPENDIX

1. A collection of particles comprising metal vanadium oxide, the particles having an average diameter less than about 1 micron.
2. The collection of particles of claim 1 wherein the particles have an average diameter from about 5 nm to about 100 nm
3. The collection of particles of claim 1 wherein the particles have an average diameter from about 5 nm to about 50 nm.
4. (Canceled)
5. (Canceled)
6. The collection of particles of claim 1 wherein less than about 1 particle in  $10^6$  have a diameter greater than about four times the average diameter of the collection of particles.
7. The collection of particles of claim 1 wherein less than about 1 particle in  $10^6$  have a diameter greater than about two times the average diameter of the collection of particles.
8. The collection of particles of claim 1 wherein the collection of particles have a distribution of particle sizes such that at least about 95 percent of the particles have a diameter greater than about 40 percent of the average diameter and less than about 160 percent of the average diameter.



9. The collection of particles of claim 1 wherein the collection of particles have a distribution of particle sizes such that at least about 95 percent of the particles have a diameter greater than about 60 percent of the average diameter and less than about 140 percent of the average diameter.
10. A method of producing particles of metal vanadium oxide comprising heating a mixture of vanadium oxide particles with a non-vanadium metal compound, the vanadium oxide particles having an average diameter less than about 1 micron.
11. The method of claim 10 wherein the vanadium oxide particles have an average diameter from about 5 nm to about 100 nm.
12. The method of claim 10 wherein the non-vanadium metal compound comprises silver nitrate.
13. The method of claim 10 wherein the vanadium oxide particles comprise crystalline  $V_2O_5$ .
14. The method of claim 10 wherein the heating is performed at a maximum temperature from about 200°C to about 330°C.
15. The method of claim 10 wherein the heating is performed at a maximum temperature from about 200°C to about 300°C.
16. The method of claim 10 wherein the heating is performed for less than about 20 hours.

17. A battery comprising a positive electrode having active particles comprising metal vanadium oxide within a binder, the active particles having an average diameter less than about 1 micron.
18. The battery of claim 17 wherein the active particles have an average diameter from about 5 nm to about 100 nm.
19. (Canceled)
20. (Canceled)
21. (Canceled)
22. The battery of claim 17 wherein the positive electrode further comprises supplementary, electrically conductive particles.
23. The battery of claim 17 wherein less than about 1 active particle in  $10^6$  have a diameter greater than about four times the average diameter of the collection of active particles.
24. The collection of particles of claim 1 wherein the particles have an average diameter less than about 500 nm.
25. The method of claim 10 wherein the vanadium oxide particles having an average diameter less than about 500 nm.

26. The battery of claim 17 wherein the active particles have an average diameter less than about 500 nm.

## **EVIDENCE APPENDIX**

A - U.S. Patent 5,549,880 to Koksang



US005349880A

# United States Patent [19]

Koksbang

[11] Patent Number: 5,549,880

[45] Date of Patent: Aug. 27, 1996

## [54] METHOD OF MAKING LITHIUM-VANADIUM-OXIDE ACTIVE MATERIAL

[76] Inventor: Rene Koksbang, 4231 Norwalk Dr.,  
San Jose, Calif. 95129

[21] Appl. No.: 221,301

[22] Filed: Mar. 31, 1994

[51] Int. Cl.<sup>6</sup> ..... C01G 31/00; H01M 4/58;  
C01D 15/02

[52] U.S. Cl. .... 423/593; 429/218

[58] Field of Search ..... 423/593; 429/218

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Primary Examiner—Charles L. Bowers, Jr.

Assistant Examiner—J. Pasterczyk

### [57] ABSTRACT

A method of making an electrode active material of the nominal general formula  $\text{LiV}_y\text{O}_z$ , where  $y$  is greater than 0 and up to about 3 and  $z$  is greater than 0 and up to about 8, comprises a series of steps. In the first step, lithium hydroxide is dispersed in an alcohol of the general formula  $\text{C}_n\text{H}_{2n+1}\text{OH}$ . The alcohol and the hydroxide are each in an amount sufficient to provide a lithium alkoxide of the general formula  $\text{LiOC}_n\text{H}_{2n+1}$ . Next, progressive amounts of an oxide of vanadium having the general formula  $\text{V}_2\text{O}_5$  (vanadium pentoxide) are added while stirring the mixture. The amount of vanadium pentoxide in the mixture is sufficient to provide about 3 moles of vanadium for each mole of lithium present in the alkoxide. Then, the mixture is heated to an elevated temperature for a time sufficient to change the color of the oxide of vanadium and provide a solid precipitate. The solid precipitate is separated from the mixture and dried to obtain a powder of an oxide of vanadium having the nominal general formula  $\text{LiV}_y\text{O}_z$ ,  $0 < y \leq 3$  and  $0 < z \leq 8$ .

16 Claims, 1 Drawing Sheet

U.S. Patent

Aug. 27, 1996

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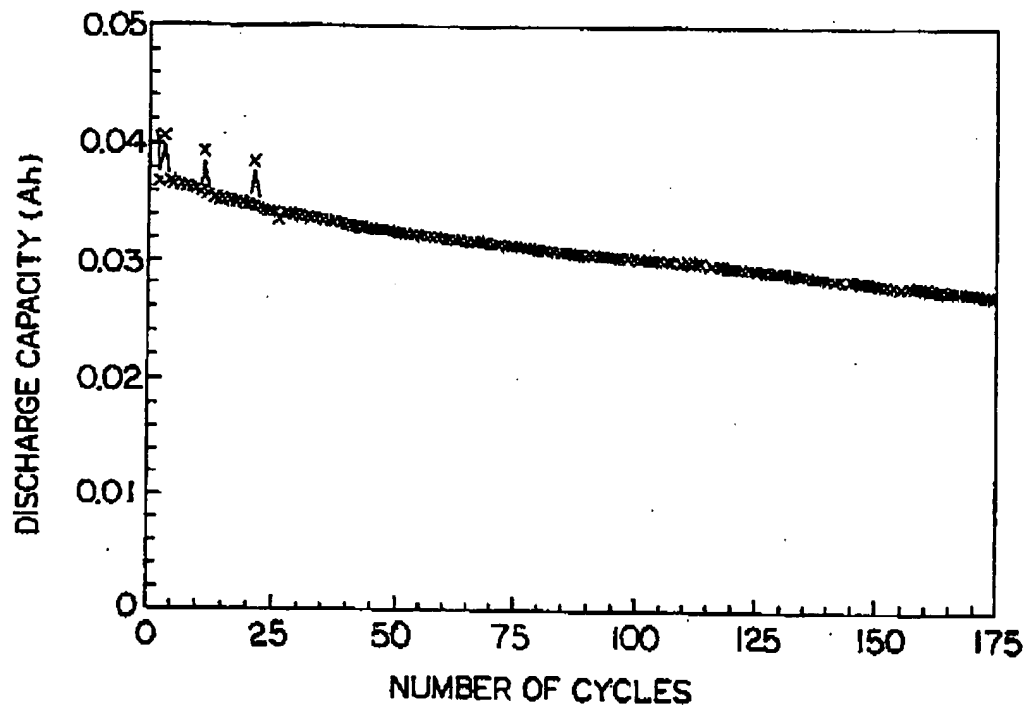


FIG. 1

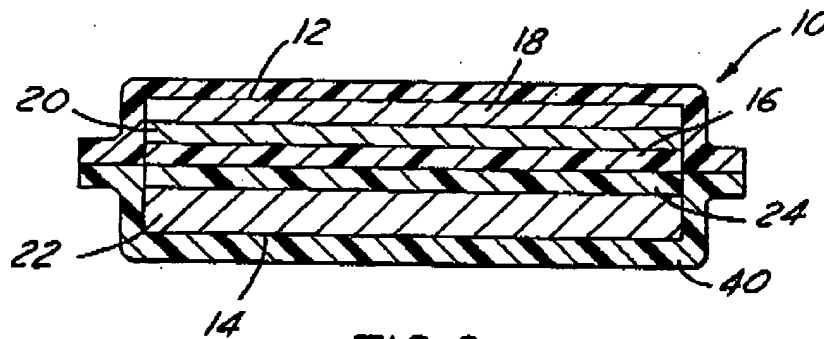


FIG. 2

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# METHOD OF MAKING LITHIUM-VANADIUM-OXIDE ACTIVE MATERIAL

## FIELD OF THE INVENTION

This invention relates to electrochemical batteries, and more particularly to improved positive electrode active material mainly composed of an oxide of vanadium.

## BACKGROUND OF THE INVENTION

Lithium-based cells or batteries often comprise cathodes of transition metal oxides which are used as intercalation compounds. The intercalation reaction involves the interstitial introduction of a guest species, namely lithium, into the host lattice of the transition metal oxide, essentially without structural modification of the host lattice. Such an intercalation reaction is essentially reversible because suitable transition states are achieved for both the forward and reverse of the intercalation reaction.

The basic components of a lithium cell typically include a lithium anode, a separator, and a metal oxide intercalation cathode active material such as a vanadium oxide compound. The cathode is usually a mixture of such an oxide compound and other components such as graphite and an electrolyte/binder which provide ionic transport. During cell operation, incorporation of lithium in the metal oxide occurs.

Current batteries contain high surface area active material such as vanadium oxide and lithium vanadium oxide powders (i.e.  $V_2O_5$ ,  $LiV_2O_5$  and  $LiV_3O_8$ ). These oxide powders are obtained, for example, by milling of vanadium oxide material. Current methods for the manufacture of powders involve mechanical grinding of vanadium oxide material prepared, for example, by rapid quench of molten material or by precipitation from an aqueous solution.

U.S. Pat. No. 5,013,620 describes solid state synthesis of  $Li_{1-x}V_3O_8$  obtained by high temperature melting (at least 700° C.) of  $V_2O_5$  with  $Li_2CO_3$ , in suitable proportions. The melt, once cooled, gives rise to solid lumps of material which are then difficult to crush and mill in order to obtain the cathode material. In addition, there is reaction between the molten  $LiV_3O_8$  product and most containers which causes contamination of the product.

Formation of an oxide of vanadium in an aqueous solution of lithium hydroxide produces a gel product which is difficult to filter and dry. The dried product is in the form of lumps which are difficult to grind.

As can be seen, present processes produce vanadium oxide in the form of lumps. By standard milling techniques it is difficult to reduce the lumps to a size less than 100 micrometers (microns) and extremely difficult to achieve closer to 10 microns. Smaller vanadium oxide particle sizes are desirable because the larger the surface area, the higher is the current drawn from a battery while the current density on the surface of the vanadium oxide active material remains low which allows high utilization of the active material. A typical coarse  $V_2O_5$  powder of 95% purity available from Fisher Scientific Company, has a median particle size of about 110 microns and a surface area of about 5 meters<sup>2</sup>/gram. Such a powder would need extensive milling.

Therefore, what is needed is a new process of forming a vanadium oxide based active material which does not produce lumps and which is readily adaptable to large scale production.

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## SUMMARY OF THE INVENTION

In accordance with the invention, a method of making an electrode active material of the nominal general formula  $LiV_yO_z$ , where y is greater than 0 and up to about 3 and z is greater than 0 and up to about 8, comprises a series of steps. In the first step, lithium hydroxide is dispersed in an alcohol of the general formula  $C_nH_{2n+1}OH$ . The alcohol and the hydroxide are each in an amount sufficient to provide a lithium alkoxide of the general formula  $LiOC_nH_{2n+1}$ . Next, progressive amounts of an oxide of vanadium having the general formula  $V_2O_5$  (vanadium pentoxide) are added while stirring the mixture. The amount of vanadium pentoxide in the mixture is sufficient to provide about 3 moles of vanadium for each mole of lithium present in the alkoxide. Then, the mixture is heated to an elevated temperature for a time sufficient to change the color of the oxide of vanadium and provide a solid precipitate. The solid precipitate is separated from the mixture and dried to obtain a powder of an oxide of vanadium having the nominal general formula  $LiV_yO_z$ ,  $0 < y \leq 3$  and  $0 < z \leq 8$ . It should be noted that the value of z may slightly exceed 8 and be on the order of 8.1 but it is less than 9; and the value of y may exceed 3 but it is less than 4. Accordingly, the nominal formula remains as above.

It is preferred that the mixture contains a stoichiometric amount of the alkoxide and pentoxide. This corresponds to about 2 moles of the alkoxide for each 3 moles of vanadium pentoxide. The stoichiometric mixture is heated to elevated temperature in a range of about 70° to about 80° centigrade, for about one to about three hours. The progress of the reaction  $LiOC_nH_{2n+1} + V_2O_5 \rightarrow LiV_yO_z$  is monitored by observing a color change from yellow/red to brown/black.

Finally, the precipitate is separated from the mixture and is dried at a temperature of about 110° centigrade. A higher temperature, such as 150° C. may be used but it is not necessary.

Alcohols which are suitable for forming the lithium alkoxide include methanol, ethanol, propanol, butanol, isopropanol, and higher alcohols. The higher alcohols are likely to react more slowly with the lithium. Ethanol is preferred. When ethanol is used, the lithium hydroxide and ethanol provide a lithium alkoxide of the general formula  $LiOCH_2CH_3$  (lithium oxyate or lithium ethoxyate) according to the reaction  $LiOH + CH_3CH_2OH \rightarrow LiOCH_2CH_3 + H_2O$ . Vanadium pentoxide is added to the lithium alkoxide in an amount to provide about 3 moles of vanadium for each mole of lithium. Next, the mixture is heated to the elevated temperature for a time sufficient to react the vanadium pentoxide with the  $LiOCH_2CH_3$  until a color change demonstrates the desired product (precipitate) has been formed. The color change is the same as described above. All other steps of the process are according to the general method described earlier. In one embodiment, the ethanol is present in a solution of 96% ethanol; about 0.5 mole of lithium hydroxide is used for each 200 milliliters of 96% ethanol; and about 0.75 mole of vanadium pentoxide is used for each 0.5 mole of lithium hydroxide.

The  $LiV_yO_z$  product of the invention is in the form of a fine powder having a surprisingly small particle size on the order of 0.1 to 5 microns, and typically less than 10 microns.

The cathode material of the invention was tested in a cell to determine the behavior of specific capacity at an increasing number of charge and discharge cycles.

The rate capability and cycling capacity was markedly improved over lithium vanadium oxide prepared by conventional methods.

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It is an object of the invention to provide a new method for preparing a lithium vanadium oxide positive electrode active material for a lithium battery. Another object is to provide a lithium battery having good charge and discharge capacity. Another object is to provide an improved electrochemical battery based on lithium which maintains its integrity over prolonged life cycle as compared to presently used batteries. Another object is to provide good conversion of the starting materials to the lithium vanadium oxide product.

These and other objects, features and advantages will become apparent from the following description of the preferred embodiments, claims and accompanying drawings.

### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows the behavior of the specific capacity at an increasing number of charge and discharge cycles.

FIG. 2 is an illustration of a cross-section of a thin battery, or cell embodying the invention.

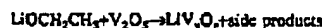
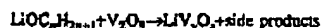
### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

#### I. Preparation of Active Material

In one embodiment a positive electrode material of the nominal general formula  $\text{LiV}_x\text{O}_y$  is prepared by a series of steps beginning with forming a mixture of lithium hydroxide dispersed in an alcohol of the general formula  $\text{C}_n\text{H}_{2n+1}\text{OH}$ . This produces a lithium alkoxide of the general formula  $\text{LiOC}_n\text{H}_{2n+1}$  according to the following general reaction:  $\text{LiOH} + \text{C}_n\text{H}_{2n+1}\text{OH} \rightarrow \text{LiOC}_n\text{H}_{2n+1} + \text{H}_2\text{O}$ .

The above reaction may be conducted under room temperature conditions, at a temperature in a range of about 15° to about 95° centigrade, and preferably 70° to 85° C. In accordance with the general formula shown above, one mole of the lithium hydroxide is reacted with one mole of an alcohol to form one mole of the lithium alkoxide product. Alcohols which are suitable for forming the lithium alkoxide include methanol, ethanol, propanol, butanol, isopropanol, and high alcohols. The higher alcohols are likely to react more slowly with the lithium. Ethanol is preferred. In the case of ethanol, the reaction to form the lithium alkoxide (lithium oxalate or lithium ethoxylate) is according to:  $\text{LiOH} + \text{CH}_3\text{CH}_2\text{OH} \rightarrow \text{LiOCH}_2\text{CH}_3 + \text{H}_2\text{O}$ .

The reaction proceeds rather quickly in about a few minutes. Once the lithium alkoxide is formed, to the mixture is added progressive amounts of an oxide of vanadium, having the general formula  $\text{V}_2\text{O}_5$  (vanadium pentoxide). The vanadium pentoxide is added progressively while stirring the mixture so as to provide about three moles of vanadium in the final product for each mole of lithium. This corresponds to a stoichiometric molar ratio of lithium alkoxide to vanadium pentoxide of about 2 to 3. This mixture is heated to a temperature higher than room temperature and preferably in a range of about 70 to about 80 degrees centigrade. Heating is conducted for at least about one half hour and preferably for about one to three hours. In order to react the lithium alkoxide with the vanadium pentoxide according to the following general formulas:



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The progress of the reaction may be followed by observing a change in color whereby the vanadium pentoxide is transformed to a lithium-vanadium oxide product and changes from a yellow-red to a brown-black. The reaction product is the brown to black solid precipitate which is separated and dried to obtain a fine powder of an oxide of the lithium-vanadium oxide having the nominal general formula  $\text{LiV}_x\text{O}_y$  where y is greater than 0 and less than or equal to about 3 and z is greater than 0 and less than or equal to about 8. The drying may occur at a temperature in a range of about 20° to about 300° centigrade. It is preferred that the drying temperature be 110° centigrade. Higher temperatures are not strictly necessary but may be more efficient.

#### Example

The method of the invention was demonstrated by using lithium hydroxide, 96% ethanol, and vanadium pentoxide powder ( $\text{V}_2\text{O}_5$ ).

Vanadium pentoxide of the general formula  $\text{V}_2\text{O}_5$  was obtained from Kerr McGee, Johnson Matthey or Alpha Products of Danvers, Mass. It had a melting point of 690° C., decomposed at 1750° C. and had a specific gravity of 3.357 grams per cc at 18° C. It was a yellow to red crystalline powder. Vanadium pentoxide has a CAS number of 1314-62-1.

Alternatively, the vanadium pentoxide may be prepared from ammonium metavanadate ( $\text{NH}_4\text{VO}_3$ ). The ammonium metavanadate is heated to a temperature of about 400° C. to about 450° C. to decompose it to vanadium pentoxide ( $\text{V}_2\text{O}_5$ ), usually in a crystalline form (in the presence of oxygen). The ammonium metavanadate is a solid crystalline material, usually a white to yellow powder. Processes for production of ammonium metavanadate are known in the art and will not be repeated here. Such processes are described in U.S. Pat. Nos. 3,063,795 and 3,063,796; and processes for preparation of ammonium metavanadate and then for production of vanadium pentoxide therefrom are described in U.S. Pat. Nos. 3,728,442, 4,061,711 and 4,119,707, each of which is incorporated by reference in its entirety.

Lithium hydroxide is a powder of essentially colorless crystals having a specific gravity of approximately 2.54 and a melting point of approximately 462° centigrade. In this example, approximately 12 grams of lithium hydroxide was dispersed in about 200 milliliters of 96% ethanol. The 12 grams of lithium hydroxide corresponds to approximately 0.5 mole.

The lithium hydroxide was dissolved in the alcohol to provide the lithium alkoxide. Next, progressive amounts of vanadium pentoxide were added to the mixture containing the lithium alkoxide while keeping the whole mixture stirred. In this example, approximately 136.5 grams of vanadium pentoxide was used which corresponds to about 0.75 mole.

The mixture was then heated to a temperature in a range of approximately 70° to 80° centigrade for about one to three hours. The progress of the reaction was monitored by a color change from the typical yellow/red  $\text{V}_2\text{O}_5$  color to a darker color which was reddish-brown to brown-black. Since monitoring of a color change may be somewhat subjective, it is sufficient to follow the color change, typically from relatively light to a relatively darker color. The color changes are dependent on several factors including the reaction chemistry, particle size, particle morphology, and impurities. Other color changes may be observed besides those described above. The product after filtering and drying,



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however, will typically appear as a brown-black color. The solid precipitate product of the invention was filtered and dried producing a fine powder of the above-mentioned brown-black color. The product was found to have a surprisingly small particle size on the order of 0.1 to 5 microns, and typically less than 10 microns.

The vanadium pentoxide is typically at least partially dissolved in the mixture. Typically, some portion of the vanadium pentoxide is dissolved and some is dispersed in the mixture. It is usually not necessary to use excess amounts of the alkoxide or the pentoxide to drive the reaction to completion. Close to stoichiometric amounts are sufficient and prevent unnecessary waste. It may become necessary in some situations to use an excess amount of the alcohol in order to keep the mixture from becoming solid before the reactions are complete. For example, a two to one ratio of alcohol to lithium hydroxide prevents the formation of a solid lump of  $\text{CH}_3\text{CH}_2\text{OLi}$ . Alternatively, it is possible to use a relatively low concentration of alcohol in water solution, i.e., 50% ethanol rather than 96% ethanol to achieve the same purpose. It is preferred to conduct the drying step in an essentially oxygen-free atmosphere such as under an inert gas, argon, helium, nitrogen, or under a vacuum or subatmospheric pressure. Although it is not thought to be necessary, it is possible to also conduct the reaction step in a similar atmosphere. It is preferred to use linear alcohols in the process, however, branched alcohols may also be used. The lower the alcohol and the less is the branching, the faster the reaction rate one expects. Ethanol is the first choice, with methanol being the second choice.

## II. Preparation of Cell Using Active Material

The cathode active material of the invention is used to prepare cathodes for lithium based electrochemical cells. The cycling performance of the active material (FIG. 1) was obtained using a cell as shown in FIG. 2. The test cell used to determine capacity had an active area of about  $28 \text{ cm}^2$ . Referring to FIG. 2, an electrochemical cell or battery 10 has a negative electrode side 12, a positive electrode side 14, and a separator 16 there-between. In accordance with common usage, a battery may consist of one cell or multiple cells. The negative electrode is the anode during discharge, and the positive electrode is the cathode during discharge. The negative electrode side includes current collector 18, typically of nickel, iron, stainless steel, and/or copper foil, and a body of negative electrode material 20. The negative electrode material 20 is sometimes simply referred to as the negative electrode or negative electrode composition. The negative electrode side 12 may consist of only a metallic electrode 20 without a separately distinguishable current collector 18. The positive electrode side 14 includes current collector 22, typically of aluminum, nickel, iron, stainless steel, and/or copper foil, or such foils having a protective conducting coating foil, and a body of positive electrode material 24. The cathode composition 24 has a typical composition as set forth in Table 1 and includes the  $\text{LiV}_2\text{O}_5$  of the invention as the active material. The positive electrode material 24 is sometimes simply referred to as the positive electrode or positive electrode composition. The separator 16 is typically a solid electrolyte or electrolyte separator. Suitable electrolyte separators (polymer electrolyte) are described in U.S. Pat. Nos. 4,830,939, 4,990,413, 5,037,712, and 5,229,225 each of which is incorporated herein by reference in its entirety. The electrolyte separator is a solid organic polymer matrix containing an ionically conducting powder or liquid with an alkali metal salt and the liquid is

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an aprotic polar solvent. Cell 10 also includes a protective covering (not shown) which functions to prevent water and air from contacting the reactive layers of the cell 10.

Cell 10 is preferably a laminar thin cell type including a lithium anode (negative electrode 20). Laminar thin-cell batteries containing lithium anodes are known in the art, and it will be appreciated that the cell can include various constructions such as bi-faced or bi-polar cell designs. Examples of cell constructions include a "jelly roll" or a fan folded laminate strip design as described in U.S. Pat. No. 4,879,190 incorporated herein by reference in its entirety.

Because the cell utilizes a lithium anode layer 20, it is necessary to manufacture the cell in a water (humidity) free environment. Lithium is extremely reactive with water and if reacted, a passivation layer can form on the surface of the anode layer, reducing the efficiency of the layer, and increasing cell impedance. Accordingly, it is particularly desirable to manufacture the cell in an environment having a relative humidity at room temperature of less than 2% (less than 300 ppm water). An environment containing between 1 ppm and 50 ppm water, and preferably less than 1 or 2 ppm water, produces a particularly efficient cell.

TABLE 1

TYPICAL CATHODE COMPOSITION	PERCENT WEIGHT
Active Material $\text{LiV}_2\text{O}_5$	45.0
Carbon	10.0
Propylene Carbonate (PC)	33.0
PolyEthylene Oxide (PEO)	1.0
PolyEthyleneGlycolDiAcrylate (PEGDA)	9.0
TriMethylPolyEthylene Oxide Tri-Acrylate (TMPEOTA)	2.0

The cathode composition containing the active material of the invention is coated onto nickel foil, followed by electron beam curing (cross-linking/polymerization) of the acrylate component. Then the electrolyte is coated on top of the cathode and cured with ultraviolet light. The lithium electrode is applied on top of the electrolyte separator and the battery is finally placed in a flexible pouch 40 which is heat sealed under vacuum.

The cathode material of the invention was tested in a cell to determine the behavior of specific capacity at an increasing number of charge and discharge cycles.

The rate capability and cycling capacity was markedly improved over lithium vanadium oxide prepared by conventional methods. The improved behavior of the specific capacity at an increasing number of charge and discharge cycles is as shown in FIG. 1.

Although not wishing to be held to any particular theory, it is thought that the product of the invention is in the form of  $\text{LiV}_2\text{O}_5$  with perhaps a small amount of  $\text{LiV}_3\text{O}_8$ . Hence, the product  $\text{LiV}_2\text{O}_5$ ,  $0 < y \leq 3$  and  $0 < z \leq 8$  encompasses both the  $\text{LiV}_2\text{O}_5$  and  $\text{LiV}_3\text{O}_8$ . The product of the invention is preferred compared to other lithium vanadium oxide particles because of the small particle size. In an as-synthesized condition the product is a powder with particle size on the order of 10 microns or less and is easily crushed to a smaller size during the formation of the cathode mixture. The fineness of the particles is important to good performance of the cathode itself because many advantages are obtained. They include no breaking of large particles which typically occurs during intercalation/disintercalation cycles; no contact loss occurs; better contact between the active material and the conductive material (carbon) so it is possible to

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apply high current; and the electrolyte can reach into innermost portions of the cathode to reduce polarization. Such advantages are evidenced by the cycling performance shown in FIG. 1.

The invention provides a lithium vanadium oxide compound having high purity, and good energy, power and cycling capability. The process of the invention is efficient and readily adaptable to continuous production of large quantities of active material in a manufacturing setting.

While this invention has been described in terms of certain embodiments thereof, it is not intended that it be limited to the above description, but rather only to the extent set forth in the following claims.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined in the appended claims.

We claim:

1. A method of making an electrode active material of the nominal general formula  $\text{LiV}_y\text{O}_z$  where y is greater than 0 but less than or equal to 3, and z is greater than 0 but less than or equal to 8, comprising:

a. forming a mixture of lithium hydroxide dispersed in an alcohol of the general formula  $\text{C}_n\text{H}_{2n+1}\text{OH}$ , each of said lithium hydroxide and said alcohol being provided in an amount sufficient to provide a lithium alkoxide of the general formula  $\text{LiOC}_n\text{H}_{2n+1}$ ;

b. adding to the mixture of step (a) progressive amounts of an oxide of vanadium having the general formula  $\text{V}_2\text{O}_5$  (vanadium pentoxide) while stirring the mixture;

c. reacting the vanadium oxide with the alkoxide in the mixture of step (b) for a time sufficient to provide a solid precipitate of the general formula  $\text{LiV}_y\text{O}_z$ ; and

d. separating and drying the solid precipitate of step (c) to obtain a powder of an oxide of vanadium having the nominal general formula  $\text{LiV}_y\text{O}_z$ , where  $0 < y \leq 3$  and  $0 < z \leq 8$ .

2. The method according to claim 1 wherein the mixture of step (b) includes 2 moles of the alkoxide for each 3 moles of vanadium pentoxide.

3. The method according to claim 1 wherein the reacting step (c) is monitored by observing color change as the precipitate is formed.

4. The method according to claim 1 wherein the reaction temperature of step (c) is in a range of 70° to 85° centigrade.

5. The method according to claim 4 wherein the reaction temperature of step (c) is maintained for one half to three hours.

6. The method according to claim 1 wherein the drying step is conducted at a temperature of 110° to 150° centigrade.

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7. The method according to claim 1 wherein the alcohol is selected from the group consisting of methanol and ethanol.

8. A method of making an electrode active material of the general formula  $\text{LiV}_y\text{O}_z$ , where y is greater than 0 but less than or equal to 3, and z is greater than 0 but less than or equal to 8, comprising:

a. forming a mixture of lithium hydroxide dispersed in ethanol each in an amount sufficient to provide a lithium alkoxide of the general formula  $\text{LiOCH}_2\text{CH}_3$  according to the reaction



b. adding to the mixture of step (a) progressive amounts of an oxide of vanadium having the general formula  $\text{V}_2\text{O}_5$  (vanadium pentoxide) while stirring the mixture;

c. reacting the oxide of vanadium with the  $\text{LiOCH}_2\text{CH}_3$  in the mixture of step (b) for a time sufficient to provide a solid precipitate of the general formula  $\text{LiV}_y\text{O}_z$ ; and

d. separating and drying the solid precipitate of step (c) to obtain a powder of an oxide of vanadium having the nominal general formula  $\text{LiV}_y\text{O}_z$ , where  $0 < y \leq 3$  and  $0 < z \leq 8$ .

9. The method according to claim 8 wherein the ethanol is present in a solution of at least 50% ethanol.

10. The method according to claim 8 wherein 96% ethanol is used and 0.5 moles of lithium hydroxide are used for each 200 milliliters of 96% ethanol.

11. The method according to claim 8 wherein 0.75 moles of vanadium pentoxide are used in step (b) for each 0.5 moles of lithium hydroxide used in step (a).

12. The method according to claim 8 wherein the mixture of step (b) includes 2 moles of the alkoxide of step (a) for each 3 moles of vanadium pentoxide.

13. The method according to claim 8 wherein reacting step (c) is monitored by observing color change as the precipitate is formed.

14. The method according to claim 8 wherein the reaction temperature of step (c) is in a range of about 70° to 85° centigrade.

15. The method according to claim 14 wherein the reaction temperature of step (c) is maintained for one half to three hours.

16. The method according to claim 8 wherein the drying step is conducted at a temperature of about 110° centigrade.

\* \* \* \* \*

**B - U.S. Patent 5,512,214 to Koksang**



US005512214A

**United States Patent** [19]

Koksang

[11] **Patent Number:** **5,512,214**[45] **Date of Patent:** **Apr. 30, 1996**[54] **LITHIUM BATTERY ELECTRODE COMPOSITIONS**[76] Inventor: **Rene Koksang**, 4231 Norwalk Dr., No. EE101, San Jose, Calif. 95129[21] Appl. No.: **324,371**[22] Filed: **Oct. 17, 1994**

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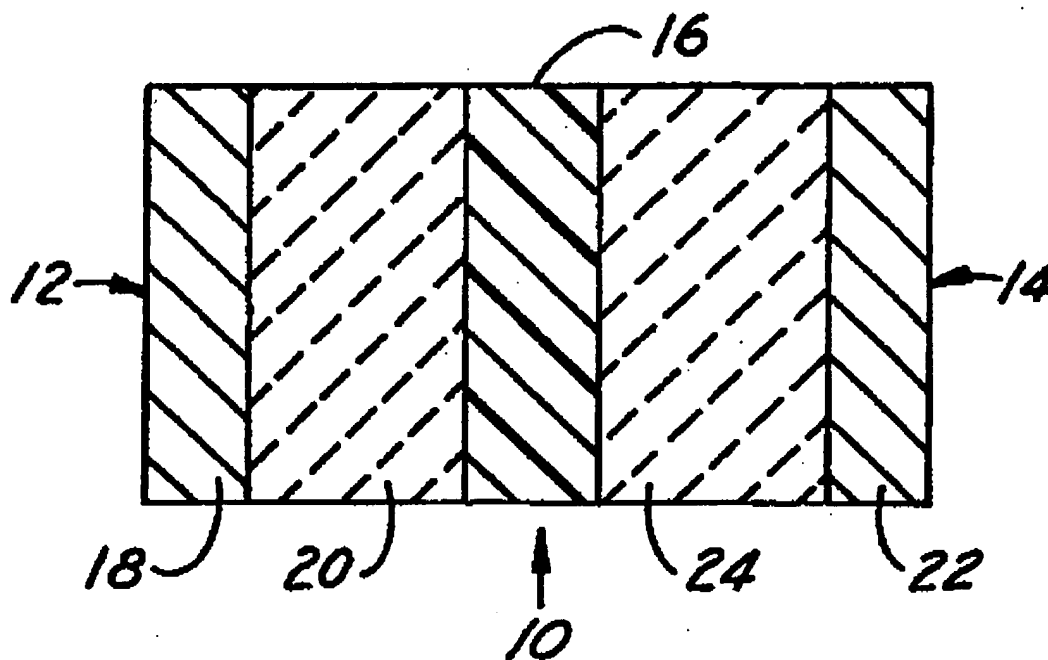
[63] Continuation of Ser. No. 40,224, Mar. 30, 1993, abandoned.

[51] Int. Cl.<sup>6</sup> ..... **H01B 1/00; H01B 1/04; H01B 1/08; H01M 4/58**[52] U.S. Cl. .... **252/506; 252/502; 252/518; 429/209; 429/212; 429/218; 429/232**[58] Field of Search ..... **252/502, 506, 252/518; 429/209, 212, 218, 232****References Cited****U.S. PATENT DOCUMENTS**

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**Primary Examiner**—Paul Lieberman**Assistant Examiner**—M. Kopec[57] **ABSTRACT**

A process of making lithium battery electrode active material having fine particles of vanadium oxide ( $V_2O_5$ ) or lithium-vanadium oxide ( $Li_xV_2O_5$ ) intimately mixed with fine particles of carbon. The process includes forming a wet solution of a vanadium oxide precursor and carbon, and then decomposing the precursor/carbon mixture at an elevated temperature or by atomization in a controlled atmosphere. Alternatively, fine particles of vanadium oxide are formed from precursor halogen compounds by atomization in oxygen.

**22 Claims, 1 Drawing Sheet**

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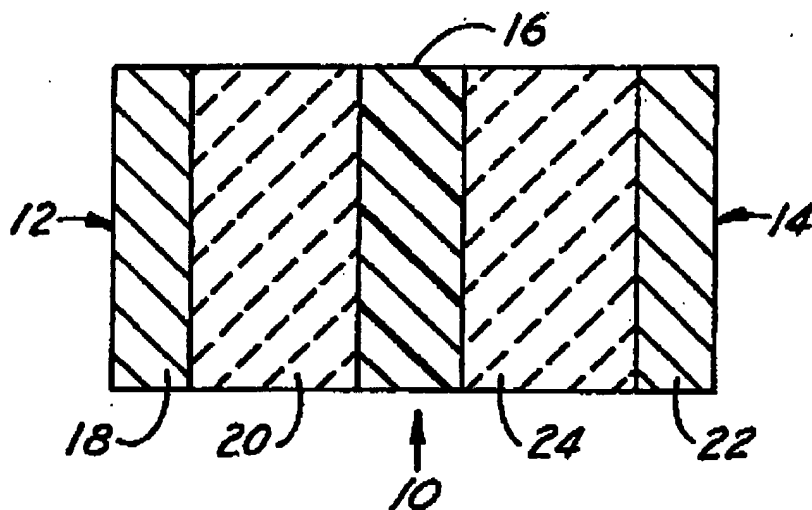


Fig. 1

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# LITHIUM BATTERY ELECTRODE COMPOSITIONS

This is a continuation of application Ser. No. 08/040,224 filed on Mar. 30, 1993 now abn.

## FIELD OF THE INVENTION

This invention relates to electrochemical batteries and more particularly to improved positive electrode material for use with lithium-containing negative electrodes.

## BACKGROUND OF THE INVENTION

The electronic conductivity of transition metal oxides, is generally lower than the electronic conductivity of transition metal sulphides. It is possible to make battery cathodes containing transition metal sulphide active material and a binder. However, it is necessary to add carbon to the composite cathode when the active material is based on an oxide. The requirement for carbon and the amount thereof depends, to some extent, on the specific oxide. The electronic conductivity of vanadium oxides decreases substantially (2-4 orders of magnitude) during lithium insertion upon discharge of a battery. This increases the need for even greater amounts of added carbon. The amount of carbon added is usually equivalent to about 40-50% by volume of the cathode mixture. This depends on the relative densities of the carbon and the other components in the cathode. Methods which allow reduction of the carbon content are important in order to increase the specific energies of the battery.

Current batteries contain high surface area oxide powders obtained by milling of precursor material. Current methods for the manufacture of powders involve mechanical grinding of precursor material prepared, for example, by rapid quench of molten material or by precipitation from an aqueous solution. As a result, the precursor material is in the form of lumps or large particles. By standard milling techniques it is difficult to reduce the lumps to less than 100 micrometers and very difficult to grind them to less than 10 to 50 micrometers. Smaller particle sizes are desirable because the larger the surface area, the higher is the current drawn from a battery while the current density on the surface of the active material remains low which allows high utilization of the active material.

## SUMMARY OF THE INVENTION

According to one aspect of the invention, there is provided a method for preparing an electrode material comprising an oxide of vanadium (vanadium oxide) and carbon, in which a solution or wet mixture comprising a compound containing vanadium oxide is prepared with fine particles of carbon dispersed therein. The solution is then dried while maintaining dispersion of the carbon particles, and then is heated for a time and at a temperature sufficient to decompose the vanadium oxide (precursor) to one or more oxides of vanadium, but insufficient to ignite the fine particles of carbon. Carbon particles are formed from precursor materials (i.e. acetylene) at temperatures of about 800° C. Thus, a suitable upper limit for the process of the invention is 800° C. However, lesser temperatures, as low as about 300° C., may be used. The decomposition may occur in minutes, depending on the temperature used. The product so formed has vanadium oxide particles intermingled with fine particles of carbon.

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Suitable precursors of the vanadium oxides are compounds selected from the group consisting of  $\text{NH}_4\text{VO}_3$ , and vanadium oxide halogen compounds such as  $\text{VOF}_2$ ,  $\text{VOBr}$ ,  $\text{VOBr}_2$ ,  $\text{VOCl}$ ,  $\text{VOCl}_2$ ,  $\text{VOCl}_3$  and similar compounds having the Br and Cl replaced by I or F. In the case of  $\text{NH}_4\text{VO}_3$ , the wet mixture is desirably aqueous; and the  $\text{NH}_4\text{VO}_3$  (ammonium metavanadate) is the precursor of the  $\text{V}_2\text{O}_5$  (vanadium pentoxide). The  $\text{V}_2\text{O}_5$  intermingled with carbon is formed by preparing an aqueous solution comprising ammonium metavanadate and fine particles of carbon dispersed in the aqueous solution. The aqueous solution is dried while maintaining dispersion of the carbon particles and then the ammonium metavanadate is decomposed to form  $\text{V}_2\text{O}_5$  intermingled with the carbon particles.

In a series of additional steps, the  $\text{V}_2\text{O}_5$  intermingled with the fine carbon particles may be included in a second wet mixture comprising  $\text{LiOH}$ . A reaction is conducted so as to form  $\text{LiV}_3\text{O}_8$  intermingled with the particles of carbon.

In the case of  $\text{VOF}_2$ ,  $\text{VOBr}$ ,  $\text{VOBr}_2$ ,  $\text{VOCl}$ , the wet mixture is desirably non-aqueous. Vanadium oxide halogens such as  $\text{VOCl}_2$ ,  $\text{VOCl}_3$  and  $\text{VOBr}$  are liquid or very deliquescent and may be mixed with carbon with or without a non-aqueous solvent.

The decomposition of the vanadium oxide compound occurs in a range of about 200° C. to about 500° C. with various compounds having various decomposition temperatures. When decomposition is conducted in an environment which is essentially oxygen-free, the oxide of vanadium so formed is represented by the nominal formula  $\text{V}_6\text{O}_{13}$ . When the decomposition is performed in an oxygen-containing environment, the oxide of vanadium produced is represented by the nominal formula  $\text{V}_2\text{O}_5$ .

When decomposition occurs in an oxygen-containing environment, so as to cause formation of  $\text{V}_2\text{O}_5$ , additional steps can be added in order to form a lithium/vanadium oxide of the nominal formula  $\text{LiV}_3\text{O}_8$  intermingled with the fine carbon particles, similar to the steps described with respect to the  $\text{NH}_4\text{VO}_3$  precursor. The additional steps include forming a second wet mixture comprising  $\text{V}_2\text{O}_5$  and lithium hydroxide and reacting the lithium hydroxide with the  $\text{V}_2\text{O}_5$  to provide the  $\text{LiV}_3\text{O}_8$  intermingled with fine particles of carbon.

Preferably, the reaction takes place at a temperature of at least about 20° C. It occurs more rapidly in a range of about 20° C. to about 60° C. It is desired that the  $\text{LiOH}$  be present in an amount sufficient to provide at least one mole of Li for each 1.5 moles of the  $\text{V}_2\text{O}_5$ .

In still another embodiment, fine particles of an oxide of vanadium are prepared by decomposition of a precursor by spray drying or atomizing. In this method, a liquid containing a vanadium oxide compound is atomized by contacting the liquid with a humidified gas stream at a temperature greater than the temperature of the liquid thereby providing very fine particles of an oxide of vanadium. Suitable vanadium oxides include vanadium oxide halogen compounds. Vanadium oxide halogen compounds which are suitable include:  $\text{VOCl}$ ,  $\text{VOCl}_2$ ,  $\text{VOCl}_3$ ,  $\text{VOBr}$ ,  $\text{VOBr}_2$ ,  $\text{VOBr}_3$  and  $\text{VOF}_2$ . These compounds are readily decomposed in humidified air to form vanadium oxides  $\text{V}_2\text{O}_5$ . Similar vanadium oxide halogen compounds having the Br, or Cl, replaced by I, or F, (x=1, 2 or 3) may also be used. Among these vanadium compounds,  $\text{VOCl}_2$ ,  $\text{VOCl}_3$  and  $\text{VOBr}_3$  are in a liquid or very deliquescent state and optionally include a solvent. In the case of the others, the liquid to be atomized includes a carrier solvent, preferably a non-aqueous, organic solvent.

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If desired, prior to atomization, the liquid containing the vanadium oxide compound may also include fine particles of carbon which are ejected into the atomized stream along with the vanadium oxide compound. This forms fine particles of vanadium oxide  $V_2O_5$ , intermingled with carbon particles.

It is an object of the invention to provide electrodes of improved specific energies by reducing vanadium oxide particle size and by improving contact between oxide particles and conductive carbon of the electrode. Other objects include reducing cost of production, reducing or eliminating milling, and increasing consistency and purity of the electrode active material.

These and other objects, features and advantages will become apparent from the following detailed description of the preferred embodiments, appended claims and accompanying drawings.

#### BRIEF DESCRIPTION OF THE DRAWING

The FIGURE is an illustration of a cross-section of a thin battery or cell embodying the invention.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

As shown in the drawing, a lithium electrochemical cell or battery 10 has a negative electrode side 12, a positive electrode side 14, and a separator 16 therebetween. In accordance with common usage, a battery may consist of one cell or multiple cells. The negative electrode is the anode during discharge, and the positive electrode is the cathode during discharge. The negative electrode side includes current collector 18, typically of nickel, iron, stainless steel, and/or copper foil, and a body of negative electrode active material 20. The negative electrode active material 20 is sometimes simply referred to as the negative electrode. The positive electrode side includes current collector 22, typically of aluminum, nickel, iron, stainless steel, and/or copper foil (such foils usually have a protective conducting coating), and a body of positive electrode active material 24 which has as its main component one or more oxides of vanadium. The positive electrode active material 24 is sometimes simply referred to as the positive electrode. The separator 16 is typically a solid electrolyte, or electrolyte separator. A suitable electrolyte separator (polymer electrolyte) is described in U.S. Pat. No. 4,830,939, incorporated herein by reference. The electrolyte separator is a solid organic polymer matrix containing an ionically conducting liquid with an alkali metal salt and the liquid is an aprotic polar solvent.

In one embodiment, positive electrode active material of vanadium oxide is intermingled with carbon and is prepared from an ammonium metavanadate precursor. The ammonium metavanadate ( $NH_4VO_3$ ) may be prepared from alkali solutions of  $V_2O_5$ , and precipitation with ammonium chloride. The starting material, ammonium metavanadate, is dissolved in solution, preferably an aqueous solution. Ammonium metavanadate is soluble in 165 parts water, and its solubility increases with temperature. Ammonia enhances solubility in water.

Next, a desired amount of carbon is dispersed in the aqueous solution. The solution is dried and in the preferred method, water is removed by evaporation while the carbon is dispersed in the solution by stirring or by agitation of the reaction vessel.

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The final step of the process consists of heating the dried mixture of carbon and ammonium metavanadate for a time and at a temperature sufficient to decompose the ammonium metavanadate to one or more oxides of vanadium, but at a temperature insufficient to ignite the fine particles of carbon. When the decomposition is conducted in an oxygen-containing environment, the vanadium oxide, produced by such decomposition, has the nominal formula  $V_2O_5$  (vanadium pentoxide). When the decomposition step is conducted in an essentially oxygen-free environment, the vanadium oxide so produced is represented by the nominal formula  $V_6O_{13}$ . Preferably, the non-oxygen containing environment is an inert gas stream, such as argon or nitrogen, whereby the  $NH_4VO_3$  is decomposed to  $V_6O_{13}$ . Thus,  $V_2O_5$  and  $V_6O_{13}$  are each formed from the decomposition product of  $NH_4VO_3$ , while being intermingled with carbon particles. The product is thus essentially in the form of carbon particles coated with the vanadium oxide or, depending on the relative sizes of the carbon particles and the vanadium oxide particles, the oxide particles may be coated with carbon. In any event, intimate mixing and intimate contact between carbon grains and oxide grains is achieved.

It is thought that the step of heating to decompose may be conducted at a temperature in the range of 400° C. to 450° C., or somewhat less because the carbon may act as a catalyst for the decomposition reaction. Thus, a temperature as low as about 300° C. may be selected. An advantage of the method is that the intimately mixed carbon and vanadium oxide is formed by heating in an atmosphere which contains  $NH_3$  formed by decomposition of the ammonium metavanadate. This is thought to be beneficial because the reducing effect of  $NH_3$  at 400° C. may remove sulfur and organic impurities present in the carbon particles. Suitable carbon particles are Ketjen Black or Shawinigan Black which are mixed with the starting  $NH_4VO_3$  prior to evaporation of the solvent and decomposition of the  $NH_4VO_3$ , as stated above. Carbon particles are formed from precursor materials (i.e. acetylene) at temperatures of about 800° C. Thus, a suitable upper limit for the process of the invention is 800° C. However, lesser temperatures, as low as about 300° C., may be used. The decomposition may occur in minutes, depending on the temperature used.

Preferably, the carbon particles are impregnated with an aqueous solution of the  $NH_4VO_3$ . Preferably, after dispersion of the carbon in the  $NH_4VO_3$  solution, the excess solvent is removed by evaporation, filtering and/or vacuum drying. Elevated temperature may be used to speed the drying process.

Carbon particles may be obtained from Noury Chemical Corporation, under the designation Ketjen Black. The Ketjen Black particles, in an as-received condition, have a BET surface area of approximately 900 m<sup>2</sup>/gram. Ketjen Black has an average particle size or equivalent average diameter in the range of about 10 to 100 nanometers (0.01 to 0.1 microns), and typically in the order of 30 nanometers. Thus, the carbon particles and oxide particles are very fine and of micron or submicron size.

The advantages of this procedure are that there is improved grain-to-grain contact between carbon and vanadium oxide particles ( $V_2O_5$  or  $V_6O_{13}$ ), and also between the various carbon particles, which enhances the electric contacts in the carbon and vanadium oxide network of the composite electrode. Another advantage is that the decomposition may occur in a temperature of about 300° C. or less, as compared to the 400° C. for decomposition of  $NH_4VO_3$  in the absence of carbon. Thus, the process of the invention reduces energy requirements and preparation costs. Still,

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another advantage is that vanadium oxide particles are formed having an average size less than 100 microns, desirably less than 50 microns and, preferably, less than 10 microns. It should be noted that median particle size refers to that size at which 50% by weight of the particles are, respectively, above and below in size.

In another embodiment, the method of the invention can be used to prepare  $\text{LiV}_3\text{O}_8$  in a completely amorphous state and in intimate contact with particles of carbon. The amorphous state is beneficial for rate capabilities and energy density. This composite is prepared by forming a  $\text{V}_2\text{O}_5$ /carbon mixture, as described above. This  $\text{V}_2\text{O}_5$ /carbon composition is then dispersed in a solution of  $\text{LiOH}$  in water, where the amount of  $\text{V}_2\text{O}_5$  and  $\text{LiOH}$  corresponds to the stoichiometric amounts of the two compounds needed for formation of  $\text{LiV}_3\text{O}_8$ . It is thought that part of the  $\text{V}_2\text{O}_5$  dissolves in solution, followed by reaction between the dissolved  $\text{V}_2\text{O}_5$ ,  $\text{LiOH}$  and water and then with the remaining solid  $\text{V}_2\text{O}_5$ , with the subsequent formation of  $\text{LiV}_3\text{O}_8$ . The reaction is fairly slow at room temperature, but proceeds at a reasonable rate at a  $50^\circ\text{C}$ . to  $60^\circ\text{C}$ . A process of adding progressive amounts of vanadium pentoxide to an  $\text{LiOH}$  solution is more fully described in U.S. Pat. No. 5,039,582, which is incorporated herein in its entirety by reference. The reaction to form  $\text{LiV}_3\text{O}_8$  is conducted while keeping the carbon and undissolved  $\text{V}_2\text{O}_5$  dispersed in solution, so that the contact between the product  $\text{LiV}_3\text{O}_8$  and the carbon is optimized. Advantageously, because part of the  $\text{V}_2\text{O}_5$  is dissolved during the procedure, intimate mixing between the  $\text{V}_2\text{O}_5$  precursor from which the  $\text{LiV}_3\text{O}_8$  is formed and the carbon, is essentially automatically achieved. Particle size of the lithium vanadium oxide is on the order of that described earlier in connection with  $\text{V}_2\text{O}_5$  and  $\text{V}_6\text{O}_{13}$ .

In still another embodiment, other vanadium oxide compounds are decomposed to form binary vanadium oxides  $\text{V}_x\text{O}_y$  intermingled with fine carbon particles. Suitable vanadium oxide compounds include vanadium oxide halogens such as  $\text{VOBr}$ ,  $\text{VOBr}_2$ ,  $\text{VOCl}$  and  $\text{VOF}_2$ , which are all thought to decompose below  $200^\circ\text{C}$ . except  $\text{VOBr}$  which decomposes at a temperature of about  $480^\circ\text{C}$ . Others are  $\text{VOCl}_2$ ,  $\text{VOCl}_3$  and  $\text{VOBr}_3$ , which are liquids and, therefore, easy to mix with carbon or absorb carbon. The family of vanadium oxide halogens which are useful in the process are represented by the general formula  $\text{V}_x\text{O}_y\text{M}_z$ , where  $x=1, 2$  or  $3$ .

In the basic method, the vanadium oxide halogen compound is mixed with particles of carbon, forming a wet mixture. The solvent is then dried while maintaining dispersion of the carbon particles, and then is heated for a time and at a temperature sufficient to decompose the vanadium oxide compound to one or more oxides of vanadium, but insufficient to ignite the fine particles of carbon. The product so formed comprises particles of one or more oxides of vanadium intermingled with fine particles of carbon. Given the presence of carbon, which acts as a catalyst, reduction of the stated decomposition temperatures is possible. It should be noted that at lower decomposition temperatures, some compounds may become less stable and may react with water and, therefore, require handling under inert gas conditions, or with a non-aqueous solvent.

In still another embodiment, finely dispersed powders of binary vanadium oxides are formed from vanadium oxide halogens ( $\text{V}_x\text{O}_y\text{M}_z$ ,  $\text{M}=\text{Cl}, \text{Br}, \text{F}$  or  $\text{I}$ ) by decomposition in a moist atmosphere by atomization. Such compounds are exemplified by the general formula  $\text{V}_x\text{O}_y\text{M}_z$ , where  $x=1, 2$  or  $3$ . Among the vanadium oxide halogen compounds,  $\text{VOCl}_2$ ,  $\text{VOCl}_3$  and  $\text{VOBr}_3$  are particularly suitable because they are liquids and a carrier solvent (dispersant) may not be required for atomization or spray drying. Other vanadium oxide halogen compounds such as  $\text{VOBr}$ ,  $\text{VOBr}_2$ ,  $\text{VOCl}$  and

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$\text{VOF}_2$ , are not typically in a liquid condition, these compounds may be dissolved or dispersed in a non-aqueous solvent and then atomized to achieve finely dispersed binary  $\text{V}_x\text{O}_y$  powders. When the decomposition takes place in open air, the major component of such decomposition will be  $\text{V}_2\text{O}_5$ . However, by adjusting the reducing properties of the atmosphere, various vanadium oxides in the form of fine particles may be obtained. Such particles are of micron or submicron size. In the absence of carbon the finely dispersed vanadium oxide powders so produced are then mixed with carbon without the necessity of ball-milling to achieve intimate contact. Alternatively, fine particles of carbon may be included in the liquid prior to atomizing. The conditions for atomization are rather broad and include forming droplets in a gas stream containing water, i.e. humidified gas stream. The conditions for decomposition of an atomized stream of VO halogen compounds ( $\text{V}_x\text{O}_y\text{M}_z$ ) are rather broad. From a practical view point, there must be sufficient humidity to permit reaction between  $\text{V}_x\text{O}_y\text{M}_z$  molecules and droplets or molecules of water. Thus, even a small amount of water in an air stream will yield some binary oxide product. As the air is made more humid, the dispersion and diffusion of water molecules result in improved contact with  $\text{V}_x\text{O}_y\text{M}_z$  molecules, and a greater yield of binary  $\text{V}_x\text{O}_y$  product. The temperature of such process must be sufficient to prevent recondensation of water and consequential reduction of water in the humid air. It is possible to conduct the atomization/decomposition at about room temperature, i.e. about  $10^\circ\text{C}$ . to about  $40^\circ\text{C}$ ., and then adjust the flow of atomized  $\text{V}_x\text{O}_y\text{M}_z$  and humid air to achieve a desired yield. The theoretical upper temperature limit is that at which decomposition of the desired binary oxide product, or carbon particles intermingled therewith, occurs. This is thought to be on the order of about  $800^\circ\text{C}$ .

The vanadium oxides of the invention were used to prepare cells with lithium-based anodes. Several cells were prepared by mixing oxide active material carbon (typical Shawinigan Black) and electrolyte/binder. The oxides were prepared with and without carbon. Thus, in some cases, carbon particles were added after fine particles of the oxide had been formed. A typical composition is as given in Table 1.

TABLE 1

Typical Cathode Composition	Percent by Weight
Vanadium Oxide	45.2%
Carbon	10.3%
Propylene Carbonate (PC)	33.3%
PolyEthylene Oxide (PEO)	1.0%
PolyEthyleneGlycolDiAcrylate (PEGDA)	8.8%
TriMethylPolyEthylene Oxide TriAcrylate (TMPEOTA)	1.5%

The cathode was coated onto nickel foil followed by electron beam curing (cross-linking/polymerization) of the acrylate component. Then the electrolyte was coated on top of the cathode and cured with ultraviolet light. The lithium electrode was applied on top of the electrolyte separator and the battery was finally placed in a flexible pouch which was heat sealed under vacuum.

The energy density of the batteries based on these new electrode materials is improved. That is believed to be achieved by decreasing the carbon content of the cathode, providing better contact between the carbon and the vanadium oxide, and keeping the carbon content at a lesser level than would otherwise be required due to the increased contact which increases the electronic conductivity allowing higher current drains, while the energy density remains essentially unchanged. Particle size is on the order of less than 100 microns, desirably less than 50 microns and, preferably, less than 10 microns.



5,512,214

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The method of the invention eliminates, or at least reduces, the need for standard milling techniques, whereby it is difficult to reduce particle size to less than about 100 microns. Since suitable particle size, less than 50 microns and, preferably, less than 10 microns is achieved by intimate mixing with carbon to form an electronically conducting carbon network with good contact to the active material on a microscopic scale, the invention avoids heavy-duty milling methods. However, if desired, milling of the final product is possible and desired for mixing of carbon with oxide particles if such carbon was not previously included. The degree of any milling is greatly reduced. That is, the mixing force of any subsequent milling step would be very much be reduced.

While this invention has been described in terms of certain embodiments thereof, it is not intended that it be limited to the above description, but rather only to the extent set forth in the following claims.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined in the appended claims.

I claim:

1. A method of preparing a composition for an electrode comprising forming a wet mixture consisting essentially of ammonium metavanadate ( $\text{NH}_4\text{VO}_3$ ) and fine particles of carbon dispersed in a solvent, removing at least a portion of the solvent and decomposing the ammonium metavanadate in an oxygen-containing environment at a temperature sufficient to form particles of an oxide of vanadium represented by the nominal general formula  $\text{V}_2\text{O}_5$  intermingled with and in grain-to-grain contact with the fine carbon particles.

2. The method according to claim 1, wherein the decomposition temperature is in a range of about 300° C. to about 450° C.

3. The method according to claim 1, wherein said at least portion of the solvent is removed from the mixture by filtration or vacuum sensation.

4. The method according to claim 1, wherein said at least a portion of the solvent is removed by heating to a temperature less than the decomposition temperature.

5. The method according to claim 1 and further comprising after said decomposition:

forming a second wet mixture comprising the decomposition product and lithium hydroxide ( $\text{LiOH}$ ), and reacting the  $\text{LiOH}$  with the decomposition product for a time and at a second temperature sufficient to provide lithium-vanadium oxide of the nominal formula  $\text{LiV}_2\text{O}_5$  intermingled with and in grain-to-grain contact with the fine carbon particles.

6. The method according to claim 5, wherein the second temperature is at least about 20° C.

7. The method according to claim 5, wherein the second temperature is in a range of about 20° C. to about 60° C.

8. The method according to claim 5, wherein the  $\text{LiOH}$  is present in an amount sufficient to provide at least one mole of  $\text{Li}$  for each 1.5 moles of the  $\text{V}_2\text{O}_5$ .

9. A method of preparing a composition for an electrode, comprising:

a) forming a mixture comprising a vanadium oxide halogen compound and fine particles of carbon dispersed in the mixture; and

b) decomposing the vanadium oxide halogen compound to form particles of one or more oxides of vanadium, and intermingling the particles of the one or more oxides of vanadium with the fine carbon particles in grain-to-grain contact therewith.

10. The method according to claim 9, wherein the mixture of step (a) includes a solvent and before step (b) at least a portion of the solvent is removed from the mixture.

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11. The method according to claim 9, wherein the vanadium oxide halogen compound is selected from the group consisting of  $\text{VOF}_2$ ,  $\text{VOBr}$ ,  $\text{VOBr}_2$ ,  $\text{VOBr}_3$ ,  $\text{VOCl}$ ,  $\text{VOCl}_2$  and  $\text{VOCl}_3$ .

12. The method according to claim 9, wherein the temperature is in a range of about 200° C. to about 500° C.

13. The method according to claim 9, wherein step (b) is conducted in an environment which is essentially oxygen-free, thereby providing the oxide of vanadium represented by the nominal formula  $\text{V}_2\text{O}_5$ .

14. The method according to claim 9, wherein step (b) is conducted in an oxygen-containing environment thereby providing the oxide of vanadium represented by the nominal formula  $\text{V}_2\text{O}_5$ .

15. The method according to claim 14 and further comprising:

forming a second mixture comprising the product of step (b) and lithium hydroxide ( $\text{LiOH}$ ), and reacting such mixture of  $\text{LiOH}$  and said product of step (c) for a time and at a second temperature sufficient to provide lithium-vanadium oxide of the nominal formula  $\text{LiV}_2\text{O}_5$  intermingled with the fine carbon particles.

16. The method according to claim 15, wherein the second temperature is at least about 20° C.

17. The method according to claim 15, wherein the second temperature is in a range of about 20° C. to about 60° C.

18. The method according to claim 15, wherein the  $\text{LiOH}$  is present in an amount sufficient to provide at least one mole of  $\text{Li}$  for each 1.5 moles of the  $\text{V}_2\text{O}_5$ .

19. The method according to claim 1 wherein the solvent removal and the decomposition are each conducted at an elevated temperature, and the solvent removal temperature is less than the decomposition temperature.

20. The method according to claim 9 wherein the mixture of step (a) includes a solvent, before step (b) at least a portion of the solvent is removed from the mixture, the solvent removal and the decomposition are each conducted at an elevated temperature, and the solvent removal temperature is less than the decomposition temperature.

21. A method of preparing a composition for an electrode, comprising:

a) forming a mixture consisting essentially of ammonium metavanadate ( $\text{NH}_4\text{VO}_3$ ) and fine particles of carbon dispersed in a solvent;

b) drying the mixture at a first temperature while maintaining dispersion of the carbon particles; and

c) heating the product of step (b) for a time and at a second temperature greater than the first temperature and sufficient to decompose the ammonium metavanadate in an oxygen-containing environment to form particles of one or more oxides of vanadium and insufficient to ignite the fine particles of carbon, and intermingling the particles of the one or more oxides of vanadium with the fine carbon particles in grain-to-grain contact therewith.

22. The method according to claim 21 and further comprising after step (c):

forming a second wet mixture comprising the product of step (c) and lithium hydroxide ( $\text{LiOH}$ ), and reacting the  $\text{LiOH}$  with the product of step (c) for a time and at a third temperature less than the second temperature and sufficient to provide lithium-vanadium oxide of the nominal formula  $\text{LiV}_2\text{O}_5$  intermingled with and in grain-to-grain contact with the fine carbon particles.

\* \* \* \* \*

**RELATED PROCEEDINGS APPENDIX**

The decision for Appeal No. 2006-0712 before the Board of Patent Appeals and Interferences is attached. This appeal involved a rejection over U.S. Patent 5,549,880 to Koksbang.

The opinion in support of the decision being entered today was not written for publication and is not binding precedent of the Board.

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

*Ex parte* XIANGXIN BI, NOBUYUKI KAMBE,  
SUJEET KUMAR, and  
JAMES T. GARDNER

Appeal No. 2006-0712  
Application No. 09/606,884

ON BRIEF

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U.S. PATENT AND TRADEMARK OFFICE  
BOARD OF PATENT APPEALS  
AND INTERFERENCES

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MAR 13 2006

PATTERSON, THUENTE, SIKAR  
& CHRISTENSEN, P.A.

Before GARRIS, WARREN, and WALTZ, Administrative Patent Judges.  
WALTZ, Administrative Patent Judge.

DECISION ON APPEAL

This is a decision on an appeal from the primary examiner's final rejection of claims 47 through 52.<sup>1</sup> The remaining claims pending in this application are claims 1, 4 through 11, 13 through 28, and 30 through 46, all of which have been allowed by the

<sup>1</sup>An amendment subsequent to the final rejection was submitted by appellants and entered by the examiner (see the amendment dated Mar. 5, 2004, entered as per the Advisory Action dated Mar. 23, 2004; Brief, page 2). We note that the word "collection" in claims 48-50 does not find antecedent basis in claim 47.

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examiner (Brief, page 2; Answer, page 2, ¶(3)). We have jurisdiction pursuant to 35 U.S.C. § 134.

According to appellants, the invention is directed to cathode compositions comprising submicron vanadium oxide particles and a binder, where these particles provide superior battery performance, especially in lithium-based batteries (Brief, page 2). Representative independent claim 47 is reproduced below:

47. A cathode composition comprising vanadium oxide particles having an average diameter from about 5 nm to about 500 nm and a binder.

The examiner has relied on Koksbang, U.S. Patent No. 5,549,880, issued on Aug. 27, 1996, as the sole evidence of unpatentability (Answer, page 3). Claims 47-52 stand rejected under 35 U.S.C. § 102(a) and (e) as anticipated by Koksbang (*id.*). For reasons stated in the Brief, Reply Brief, and below, we reverse the rejection on appeal.

#### OPINION

The examiner finds that Koksbang discloses secondary lithium batteries comprising a "lithiated vanadium oxide cathode active material," a lithium metal anode, and a polymer electrolyte or solid electrolyte separator, where the vanadium oxide particles are

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"in the form of a fine powder having a surprisingly small particle size on the order of 0.1 to 5 microns" (Answer, page 3).

The initial burden of establishing unpatentability, on any ground, rests with the examiner. See *In re Oetiker*, 977 F.2d 1443, 1445, 24 USPQ2d 1443, 1444 (Fed. Cir. 1992). As correctly argued by appellants (Brief, pages 8-10; Reply Brief, pages 2-3), Koksbang does not disclose or suggest that the range of particle sizes taught is an average size or diameter as required by claim 47 on appeal and the examiner has not convincingly established that the disclosure of Koksbang should be interpreted or construed as an "average" size or diameter (Answer, page 4). The examiner has cited the different methods of preparation taught by Koksbang as evidence that it is "reasonable" to interpret the range taught by the reference as a range of average particle sizes, which thus overlap with the claimed range (*id.*). This evidence is not convincing for the following reasons. As correctly argued by appellants (Reply Brief, page 2), there is no disclosure or suggestion in Koksbang that the variation in reaction starting materials or parameters would alter the product properties, e.g., the particle sizes of the product (see col. 4, ll. 15-65). Furthermore, Koksbang specifically teaches the criticality of the "particle size" of the product, disclosing a range of particle

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sizes but never disclosing or suggesting an average of particle sizes or diameters (col. 2, ll. 59-61; col. 5, ll. 1-6; and col. 6, ll. 56-60). We note that the examiner has not submitted any substantive evidence that the term "particle size" was known in this art to mean an average particle size.

The examiner has found that the vanadium oxide particles disclosed by Koksang are in the form of a fine powder with a particle size "on the order of 0.1 to 5 microns" (Answer, page 3). However, the examiner admits that Koksang discloses "a lithiated vanadium oxide cathode active material" (Answer, page 3), and Koksang only discloses particle sizes for the lithium vanadium oxide product (col. 2, ll. 59-61; col. 5, ll. 1-6; and col. 6, ll. 56-61). We find no disclosure in Koksang of any particle size for the vanadium oxide per se (e.g., see col. 4, ll. 15-40). We have construed the term "vanadium oxide particles" as found in claim 47 on appeal with "the broadest reasonable meaning of the words in their ordinary usage as they would be understood by one of ordinary skill in the art," taking into account any enlightenment of the term in the specification. *In re Morris*, 127 F.3d 1048, 1054, 44 USPQ2d 1023, 1027 (Fed. Cir. 1997). Appellants' specification discloses vanadium oxide nanoparticles per se, as well as the production of only vanadium oxide (see Figures 5-12; specification,

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page 4, ll. 19-21; page 5, l. 18-page 6, l. 17; and page 14, l. 8 et seq.). Although the transitional term "comprising" opens claim 47 on appeal to other elements or components,<sup>2</sup> we determine that the claimed "vanadium oxide particles," as understood by one of ordinary skill in this art and consistent with the specification, does not encompass other materials such as intercalated lithium.<sup>3</sup>

For the foregoing reasons and those set forth in the Brief and Reply Brief, we determine that the examiner has failed to establish a prima facie case of anticipation in view of Koksbang. Therefore we cannot sustain the rejection on appeal.

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<sup>2</sup>See *Vehicular Techs. v. Titan Wheel Int'l, Inc.*, 212 F.3d 1377, 1383, 54 USPQ2d 1841, 1845 (Fed. Cir. 2000) ("A drafter uses the term 'comprising' to mean 'I claim at least what follows and potentially more.'").

<sup>3</sup>See related Appl. No. 09/246,076, now U.S. Patent No. 6,225,007 B1, issued May 1, 2001.

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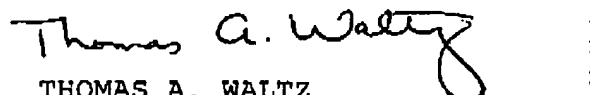
The decision of the examiner is reversed.

**REVERSED**

  
BRADLEY R. GARRIS  
Administrative Patent Judge

  
CHARLES F. WARREN  
Administrative Patent Judge

BOARD OF PATENT  
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THOMAS A. WALTZ  
Administrative Patent Judge

TAW/sld



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